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1 **Soft and flexible conductive PDMS/MWCNT composites**

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7 **Abstract**

8 Conductive elastomers based on MWCNT in polydimethylsiloxane (PDMS) have been prepared by
9 a range of dispersion methods such as ultrasonication, speedmixing and roll milling in combination
10 with physical or covalent modification. The ionic liquid (IL), 1-ethyl-3-methylimidazolium
11 bis(trifluoromethanesulfonyl)imide, was used to pre-disperse MWCNT in a MWCNT/IL-gel that
12 was used for preparation of MWCNT/PDMS composites. The method was seen to be effective at
13 low levels of MWCNT, but required combination with a roll mill to obtain a stable dispersion at
14 4 wt% MWCNT. With higher amounts of MWCNT a reduction in conductivity was observed,
15 which was attributed to a change in morphology occurring between 4 and 5wt% MWCNT. As an
16 alternative to IL dispersing aids a novel functionalized MWCNT was prepared by free radical
17 polymerization using α -methacryloxypropyl-polydimethylsiloxane, which could be used directly
18 for preparation of MWCNT/PDMS composites. Composites prepared by use of the IL dispersion
19 method, use of a roll mill or by use of the f-MWCNT all had conductivities around 0.005-0.01 S/cm
20 and retained conductivity upon extension.

21 **Introduction**

22 Conductive elastomers have been a topic of general research interest for a long time and are also
23 commercially available. They are most commonly elastomers filled with conductive particles such
24 as silver, copper, aluminium, nickel, carbon black particles or combinations thereof. Common
25 applications are e.g. electromagnetic screening materials, as flexible electrodes in sensors or in
26 flexible transducers. In particular applications of conductive silicones (polydimethylsiloxane,
27 PDMS) as compliant electrodes for dielectric elastomer (DE) transducers as well as stretchable
28 electronics¹ have received an increased attention recently². The conductive elastomer must be able
29 to sustain large deformations while remaining conductive as well as remain mechanically stable for
30 millions of cycles. Current commercial solutions cannot be used as a starting point, since these are
31 generally filled to such an extent that adding additional fillers to increase conductivity further is not
32 practically possible.

33 Several methods have been used to prepare conductive elastomers, such as grafting conductive
34 polymers to soft block copolymers³, blending conductive polymers with elastomers⁴ or fabricating
35 stretchable conductors from bacterial cellulose⁵. The preparation of these conductive elastomers is
36 either tedious and/or requires considerable amounts of solvents, which is not preferable from an
37 industrial point of view.

38 Specifically for PDMS the classical approach of adding conductive particles above the percolation
39 threshold has been investigated in several cases. Various fillers have been used in this respect, such
40 as silver nanowires (AgNWs)^{6,7}, exfoliated graphite (EG⁸) and carbon nanotubes (CNTs).

41 AgNWs were synthesised through the reduction of silver nitrate in the presence of
42 polyvinylpyrrolidone in ethylene glycol^{9,10}, resulting in a suspension that was used for elastomer
43 preparation. The prepared elastomers containing the AgNWs was shown to have a high conductivity,
44 though the conductivity was shown to decrease when the elastomer was strained^{6,7}.

45 Similarly, CNTs having single or multiple graphene layers rolled into cylinders as single walled
46 carbon nanotubes (SWCNT) or multi walled carbon nanotubes (MWCNT)^{11–13} have received much
47 attention because of their excellent mechanical, thermal and electrical properties^{14,15}. CNTs are
48 generally interesting as nanofillers for conductive elastomers, since their percolation thresholds are
49 generally very low and conductive networks are achieved at concentrations below 1 wt%, due to their
50 high aspect ratio^{12,15}. However, a drawback of CNTs is their poor dispersion in elastomers, due to the
51 formation of agglomerates as a result of the strong van der Waals force of interaction between the
52 CNTs^{16,17}. In order to achieve a better dispersion of CNTs in elastomers, covalent and non-covalent
53 modifications can be used. Covalent modification includes, amongst others, grafting functionality to
54 the CNT surface^{18–20}, while non-covalent modification usually involves mixing with a low molecular
55 weight compound such as e.g. ionic liquids (ILs) or other compatibilizers. A broad range of ILs are
56 currently available as either organic-inorganic salts or as purely organic salts and can be tailored to
57 specific properties based on the choice of cation and anion such as 1-allyl-3-methyl imidazolium, 1-
58 ethyl-3-methylimidazolium or tetralkyl ammonium in combination with
59 bis(trifluoromethylsulphonyl)imide, chloride or e.g. tetrafluoroborate. The use of ILs has generally
60 been observed to improve the dispersion of CNT in polymers, where in particular the imidazolium
61 based ILs have resulted in composites with an increased conductivity^{13,21–29}.

62 For PDMS, Oh et al.²² recently demonstrated how the use of an IL as a dispersing agent resulted in
63 well dispersed SWCNT/PDMS composites. Their focus was on development of soft systems with a
64 low amount of SWCNT (1.6 wt%) and they used large amounts of IL in order to use it both as a
65 dispersing agent as well as a plasticizer to obtain softer materials. They did not report the
66 conductivities obtained by their process, but their composites were clearly above the percolation
67 threshold.

68 In addition to effective dispersion aids, effective mixing methods are required in order to disperse
69 carbon materials in PDMS. Previous investigations in our group showed the effect of different
70 methods of dispersion such as mechanical mixing, ultrasonication and high speed mixing in a
71 Speedmixer and how that influenced the quality of an expanded graphite/PDMS composite³⁰. From
72 the available techniques in particular speedmixing and the use of a roll mill would be suitable methods
73 for use in industrial production to prepare high quality composites.

74 The purpose of this study was to identify an industrially viable process for preparation of a soft flexible
75 conductive PDMS composite with a lower degree of filling compared to commercially available
76 materials. Therefore several different approaches using different dispersing aids or mixing methods were
77 tested on an industrial grade of MWCNT.

78 **Experimental**

79 **Materials and methods**

80 MWCNTs (NC7000) were purchased from Nanocyl S.A. (Belgium). The MWCNTs is reported by
81 the supplier to have an average diameter of 9.5 nm, an average length of 1.5 μm and a surface area of
82 250-300 m^2/g . The ionic liquid, 1-ethyl-3-methylimidazolium bis(trifluoromethanesulfonyl)imide
83 and heptane were acquired from Sigma Aldrich. Elastosil RT625 and the inhibitor PT-88 were
84 purchased from Wacker Chemie AG (Germany). Elastosil RT625 is a two-component system, in
85 which component A consists of vinyl-terminated PDMS and a crosslinker, and component B consists
86 of vinyl-terminated PDMS and a catalyst, among other components such as fillers and additives. The
87 two components are mixed in a 9:1 ratio (A:B). In order to prepare the samples the zero volt ionizer
88 from Desco Industries Inc., an ultrasonic device UP200S from Heilscher, a mechanical mixer
89 Eurostar from IKA Labortechnik, a rotary-evaporator Laborota 4003 from Heidolph, a SpeedmixerTM
90 from Flack Tek Inc. and a three-roll mill from EXAKT Advanced Technologies GmbH have been

91 used. Thermo gravimetric analysis (TGA) was performed in a nitrogen atmosphere on a Q500 from
92 TA instruments with a heating rate of 10°C/min from RT to 800°C. Fourier Transform Infrared
93 spectroscopy (FT-IR) was performed on a Thermo-Fischer is50 FT-IR with a universal attenuated
94 total reflection (ATR) sampling accessory on a diamond crystal. The rheological measurements were
95 carried out on a controlled stress rheometer (AR2000) from TA Instruments at 20°C using samples
96 with a diameter of 25 mm and a thickness of 1 mm.

97 **Procedures**

98 **General procedure for preparation of MWCNT/PDMS composites**

99 For samples with IL as dispersion aid, MWCNTs and the IL, 1-ethyl-3-methylimidazolium
100 bis(trifluoromethanesulfonyl)imide, in the desired ratio were weighed in a mortar and ground with a
101 pestle until gelation was observed. Either pristine MWCNTs or the MWCNT/IL-gel was subsequently
102 dispersed in heptane at 1 mg MWCNT/mL and ultrasonicated for 14 minutes in 2 minute intervals.
103 After ultrasonication, component A of Elastosil RT625 was added and the mixture was stirred for 6
104 hours at 900 rpm. The heptane was evaporated from the mixture using a rotary-evaporator. Thereafter
105 component B of Elastosil RT625 was added and the mixture was speed-mixed for 5 minutes at
106 2750 rpm. The composite was cast onto a glass plate with a 1 mm thick frame, where after another
107 glass plate was placed on top of the sample, thus sandwiching the sample. The glass plates were
108 clamped together and the sample was cured in the oven for 10 minutes at 115 °C, resulting in a fully
109 cured PDMS/MWCNT/IL composite.

110 **Roll mill dispersion - General procedure for preparation of MWCNT/PDMS composites**

111 MWCNTs or MWCNT/IL gels, as well as both Elastosil RT625 components together with 1 wt%
112 inhibitor with respect to the PDMS (to delay the curing process), were weighed into the speed mixing
113 cup. The mixture was then speed-mixed for 5 minutes at 2750 rpm and was ready to be dispersed in
114 the roll mill. The gap between the first and second rolls was set to 30 µm, while the gap between the

115 second and third rolls was set to 15 μm and the speed of the third roll was set to 100 rpm. The sample
116 was roll-milled six times until the mixture was uniformly blended and finally coated and cured
117 thermally for 10 minutes at 115°C.

118 **Poly(MPDMS) grafted MWCNT by free radical polymerization**

119 The MWCNTs (1.5 g) were dispersed in a mixture of anisole (27 mL) and toluene (220 mL) by
120 ultrasonication for 10 min. The monomer α -methacryloxypropyl-polydimethylsiloxane (MPDMS,
121 $M_n=700$ g/mol, 33 mL, 31.8 g) and azobisisobutyronitrile (AIBN, 0.045 g, 2.4 mmol) were added to
122 the dispersed MWCNT and the reaction mixture was bubbled with nitrogen for 20 min and thereafter
123 heated to 65°C. The mixture was heated for 18 h under nitrogen and the product was isolated by
124 precipitation into MeOH (3 L), followed by filtration using a 0.4 μm PTFE filter, resulting in a black
125 viscous solid in a quantitative yield. The raw product could be used directly for preparation of
126 composites or rinsed for free polymer by washing with toluene and drying in vacuo. The raw product
127 was speed-mixed with both components of the PDMS (A and B) at 2750 rpm for 5 minutes. The
128 sample was coated on a glass plate and cured in an oven for 10 minutes at 115°C.

129 IR (cm^{-1}): 3100-2800 (CH stretch); 1728 (CO stretch); 1255 (CO stretch); 1100 and 1015 (Si-O
130 stretch) and 787 (Si-C stretch).

131 **Results and discussion**

132 The process developed by Oh et al.²² for SWCNT was adopted to higher amounts of an industrial
133 grade MWCNT to investigate it as a pathway towards highly conductive soft elastomers. The
134 preparation process is outlined in Figure 1.

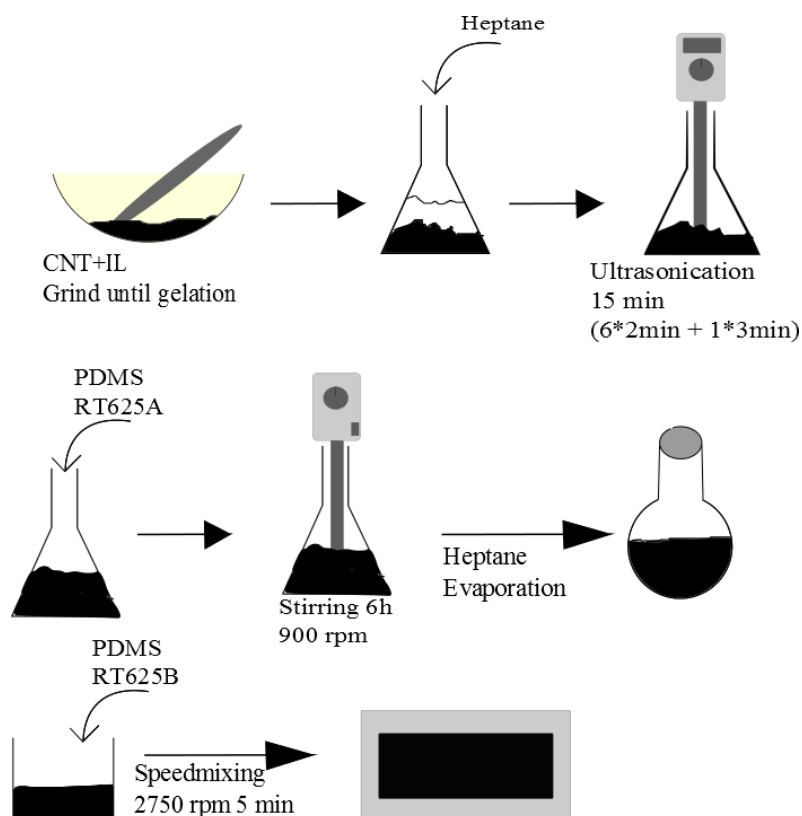


Figure 1: Schematic overview of the dispersion procedure using IL to disperse MWCNT in PDMS followed by coating and thermal curing of the MWCNT/PDMS composites.

The initial grinding of the MWCNT together with the IL (1-ethyl-3-methylimidazolium bis(trifluoromethanesulfonyl)imide) resulted in a thick gel (MWCNT/IL-gel), which was then diluted with heptane and sonicated to provide a liquid dispersion of MWCNT. Since PDMS is fully soluble in heptane this allowed direct mixing of the liquid PDMS pre-polymer into the MWCNT dispersion. This was additionally sonicated, stirred and speedmixed to provide a fully dispersed PDMS mixture that could ultimately be cured by thermal curing. The process applies a combination of dispersion methods in the form of the IL-gel, ultrasonication and speedmixing to obtain a stable dispersion of the MWCNT in PDMS.

The conductive properties of the prepared elastomers are directly linked to the quality of the dispersion and dielectric spectroscopy (DRS) was therefore used to determine to assess the quality of the dispersion as shown in Figure 2-left.

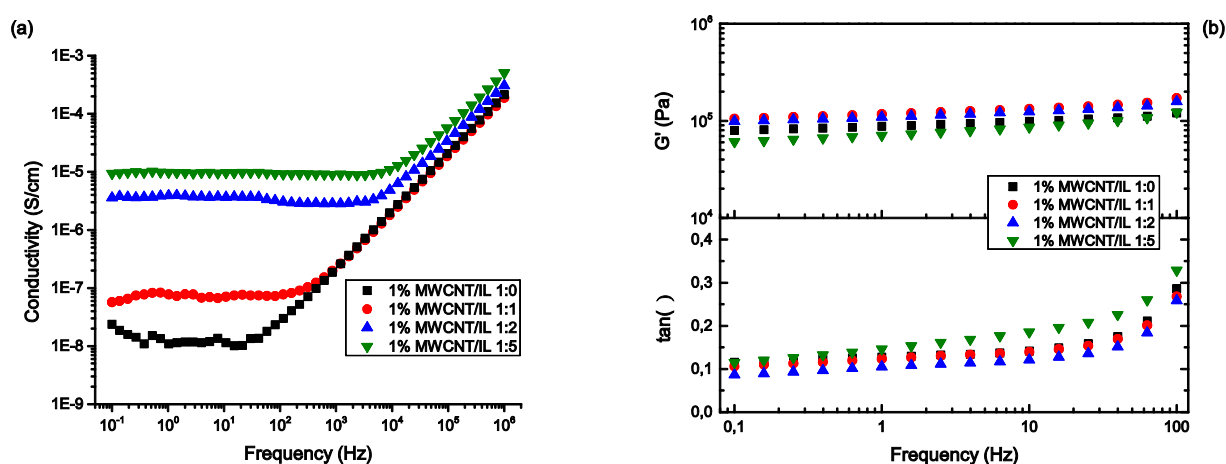
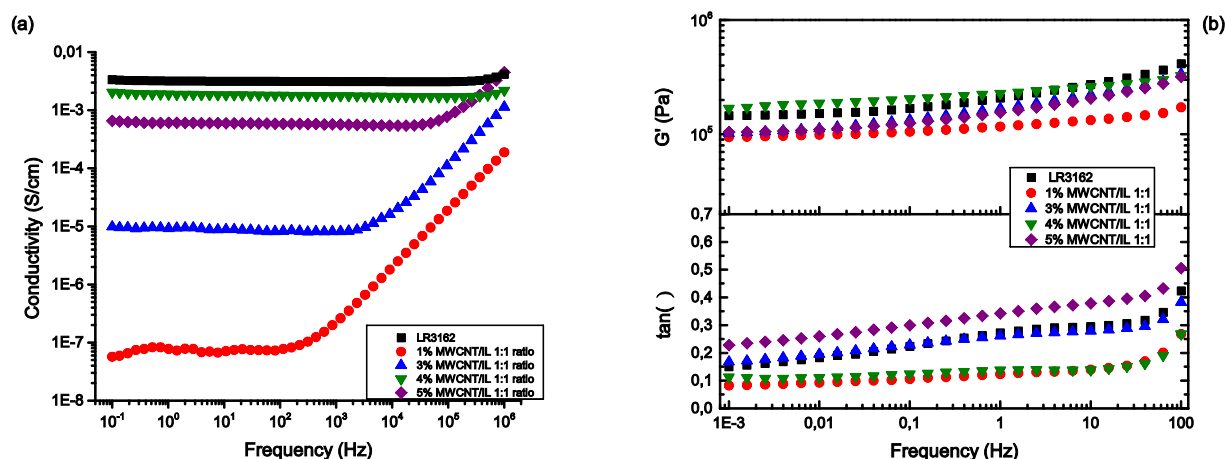


Figure 2: 1 wt% MWCNT/PDMS composites with a MWCNT/IL ratio from 1:0 to 1:5. a) Conductivities at 20°C; b) Linear viscoelastic properties at 20°C.

As shown in Figure 2a the PDMS/MWCNT composites all have a direct current plateau at lower frequencies, which is indicative for conductive materials. The level of the plateau, corresponding to the conductivity, increased with increasing amounts of IL. This is a result of an improved dispersion of the MWCNT in the matrix, since the IL is well incorporated (samples are unchanged after a year) and does not lead to a higher conductivity of the matrix, as can be seen from a reference experiment using only IL and the Elastosil 625 (SI-Figure 1). One of the disadvantages of using ILs as dispersing aids would be the cost of material as well as the impact on network integrity, when the loading becomes significantly higher than the MWCNT. It is well known that foreign compounds will leach out of the PDMS over time and the stability of the matrix was therefore investigated by rheology as shown in Figure 2b. Here it can be seen that increasing the amounts of IL relative to the MWCNT results in similar rheological behaviour, though the highest ratio tested (MWCNT/IL 1:5) shows a minor increase in $\tan(\delta)$ at higher frequencies. Generally, all of the composites show a good mechanical integrity and an effective dispersion, though increasing the MWCNT/IL ratio to 1:5 can be seen to be of negligible effect with respect to conductivity.

167 In order to increase conductivity of the composites a range of samples with higher amounts of
 168 MWCNT were prepared as shown in Figure 3.



169 **Figure 3:** MWCNT/IL PDMS composites with 1-5 wt% MWCNT at a MWCNT/IL ratio of 1:1. a) Conductivities at
 170 20°C; b) Linear viscoelastic properties at 20°C.

171

172 Increasing the content of MWCNT in the composites resulted in an effective increase in
 173 conductivity up to a loading of 4 wt% MWCNT (Figure 3a). At this point an upper limit of effective
 174 incorporation was reached as is often seen for nanocomposites, where increasing the loading further
 175 resulted in a decreased conductivity. This illustrates the necessary balance between loading,
 176 effectiveness of the filler, as well as the degree of dispersion that is critical for an effective
 177 nanocomposite. The mechanical stability of the materials was also investigated for higher amounts
 178 of MWCNT as shown in Figure 3b. With increased content of MWCNT and IL in the network,
 179 viscous losses increase for some of the systems, though this phenomenon is not reproducible and is
 180 attributed to small agglomerates resulting from minor sample to sample variations.

181 Based on the results above it can be seen that the MWCNT/PDMS composites approach the
 182 conductivity of the commercially available LR3162, which contains 40 wt% carbon black. In an
 183 effort to increase the dispersion and thereby the conductivity even further composites using a
 184 mixing ratio of MWCNT/IL 1:2 were also investigated as shown in Figure 4

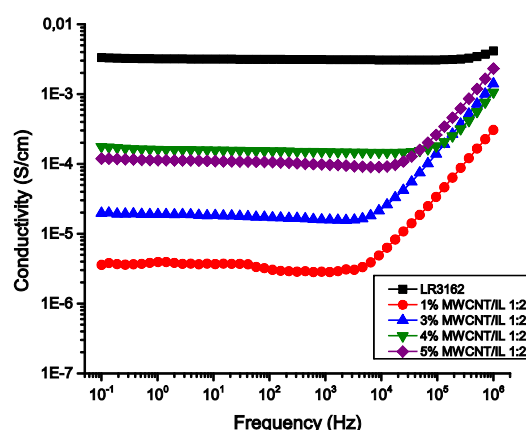


Figure 4: Conductivities of MWCNT/IL PDMS composites with a MWCNT content from 1-5 wt% with a MWCNT/IL ratio of 1:2 at 20°C.

Here it is clear that the system is not sufficiently robust at high levels of MWCNT. The conductivity increases for the 1 and 3 wt% MWCNT composites with a mixing ratio of 1:2 compared to the 1:1 mixing ratio shown above. However, increasing either the amount of MWCNT to 4 or 5 wt% or the amount of IL (compared to 1:1) does not result in an effective dispersion of the MWCNT. The upper limit observed in conductivity for the composites with 4-5 wt% MWCNT and varying amounts of IL indicates that there is a change in dispersion occurring at this specific loading. To investigate this change, a range of composites with varying IL content and high amounts of MWCNT (4-5wt%) was investigated by scanning electron microscopy (SEM) as shown in Figure 5.

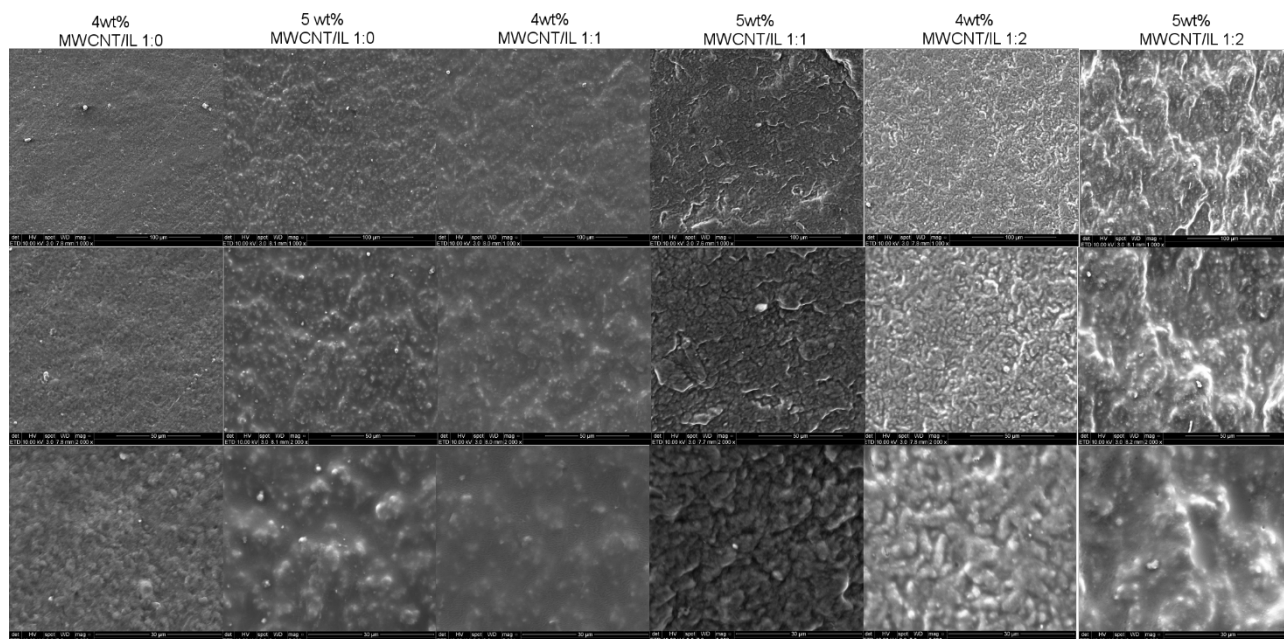


Figure 5: SEMs of the composites containing 4-5wt% MWCNT and 0-2 wt% IL. The particles (white spots) seen in the SEMs are silica particles from the Elastosil RT625.

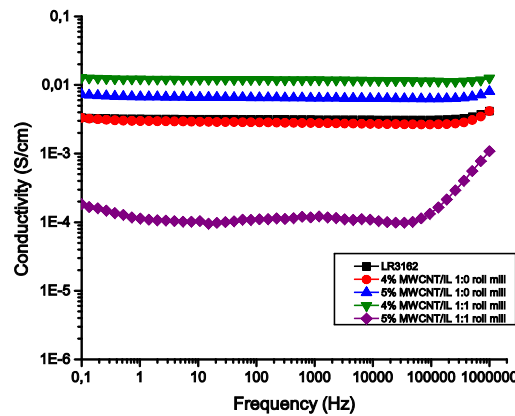
From the micrographs it is clear that samples without IL and 4-5 wt% MWCNT have a homogeneous appearance, where the MWCNT appears well distributed. With addition of a small amount of IL the picture is unchanged for 4 wt% MWCNT and the elastomer appears very similar. However, the 5 wt% of MWCNT/IL 1:1 shows a distinctly different morphology. With a higher amount of IL (MWCNT/IL 1:2) this morphology is observed for all the samples. In addition to this the samples begin to accumulate charge, which is normally observed for MWCNT composites below the percolation threshold³¹. It is unclear exactly what is happening at this composition, but it directly correlates to the low conductivities observed for these samples both for the 5wt% MWCNT/IL 1:1 and for MWNCT/IL 1:2 elastomers.

These results thereby show that moderate amounts of IL is quite effective as a dispersing aid at low amounts of MWCNT, whereas larger amounts of IL or higher amounts of MWCNT results in a less efficient dispersion.

Roll mill dispersion

Due to the good results on 4 wt% MWCNT (MWCNT/IL 1:1) this system was used to investigate the effect of using roll milling as an additional dispersion method. The process outlined in Figure 1 was additionally followed by a final treatment on a roll mill to homogenize the samples. To see the effect of using the roll mill both samples with and without IL were investigated as shown in Figure 6.

226



227

Figure 6: Conductivities of 4 and 5 wt% MWCNT/PDMS composites without and with IL dispersed by speedmixing followed by a roll mill treatment measured at 20°C.

230

Here it is clear that roll milling in combination with speedmixing provides a very efficient dispersion method. For all the roll milled samples the direct current plateau has a much earlier onset compared to the MWCNT/IL or pristine MWCNT composites. This clearly underlines that the roll mill provides a much more effective dispersion compared to the earlier mentioned methods. In the roll milled MWCNT composites without IL there is not much difference between the 4 and 5 wt% MWCNT composites. Both samples are well dispersed and provide a high conductivity comparable to that of LR3162. In contrary to this, for the samples that were pre-dispersed as an IL-gel with 4 and 5 wt% MWCNT a difference of several orders of magnitude in conductivity was seen. The 4 wt% sample can be seen to give a slightly higher conductivity than LR3162, whereas the 5 wt%

sample is clearly above the higher loading limit. The effects on the mechanical properties of the soft elastomer films were investigated by rheology as shown in Figure 7.

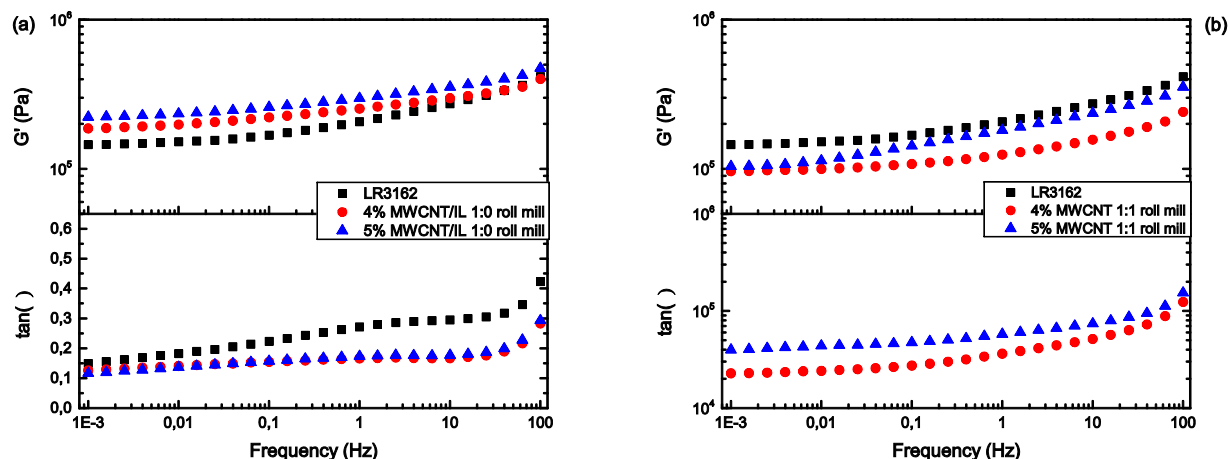


Figure 7: Linear viscoelastic properties of the roll milled 4 and 5 wt% MWCNT/PDMS composites without (a) and with (b) IL measured at 20°C.

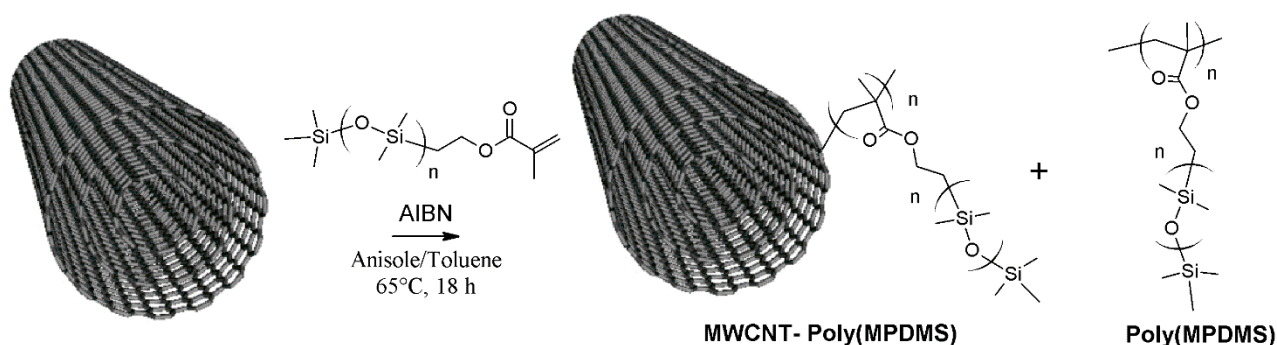
For the samples without IL (Figure 7a), G' increases in line with increasing concentration of MWCNTs and the samples are stiffer than the commercially available Elastosil LR3162. However, when the MWCNTs are pre-dispersed using the IL the composites become softer (Figure 7b). The expected influence of MWCNT on a polymer matrix would be an increase in G' as a function of loading, as e.g. observed on thermoplastic systems such as PP/MWCNT composites.³¹ For elastomers there is an additional effect from the formation of the network, where an improved dispersion influences network integrity and thereby affects the mechanical stability, which has to be taken into consideration in the design of the elastomer composition.³² For elastomers, the pristine MWCNT affects the network to an insignificant degree and results in only a moderate increase in G' . In contrast to this, the addition of the IL in combination with the improved efficiency of the dispersion by the roll mill, results in a significant influence on the mechanical properties. In particular for the 5 wt% sample the softening effect of the IL is clear, where a significant increase in

tan(δ) can be observed at low frequencies indicating a much weaker network and significant viscous loss in the network.

In general, the additional step of using the roll mill resulted in a more robust procedure that provided a very efficient dispersion. However, the positive effects of using the IL in the pre-dispersion step is counterbalanced with a significant influence on the network that is not desirable. In addition to this, the added cost of the procedure and the only moderate increase compared to the pristine MWCNT does not make this procedure competitive.

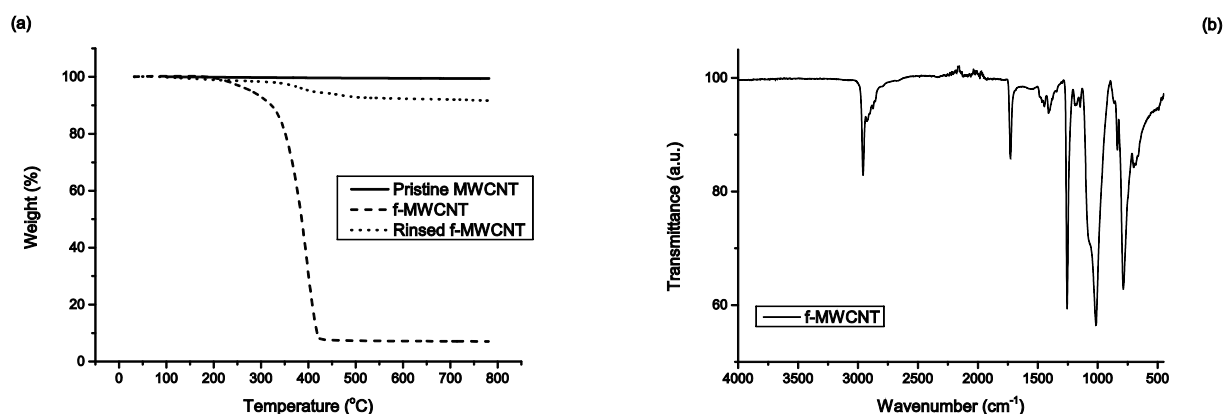
Functionalized MWCNT as a dispersion method

In addition to the use of mechanical mixing methods functionalization of nanomaterials are a well-known pathway towards improved dispersion in nanocomposites. However, as underlined by Grady³³, the use of functionalized materials will be a balance between improved dispersion and loss of MWCNT structure due to the functionalization, which ultimately results in a lower ultimate conductivity as well as a lower ultimate tensile strength of the MWCNT. Therefore a simple preparation method affording a low degree of modification as well as a product that could easily be used in a subsequent PDMS network curing reaction was developed as illustrated in Scheme 1.



Scheme 1: Synthesis of poly(MPDMS) grafted MWCNT (f-MWCNT) by in-situ free radical polymerization of MPDMS resulting in a f-MWCNT in excess free poly(MPDMS).

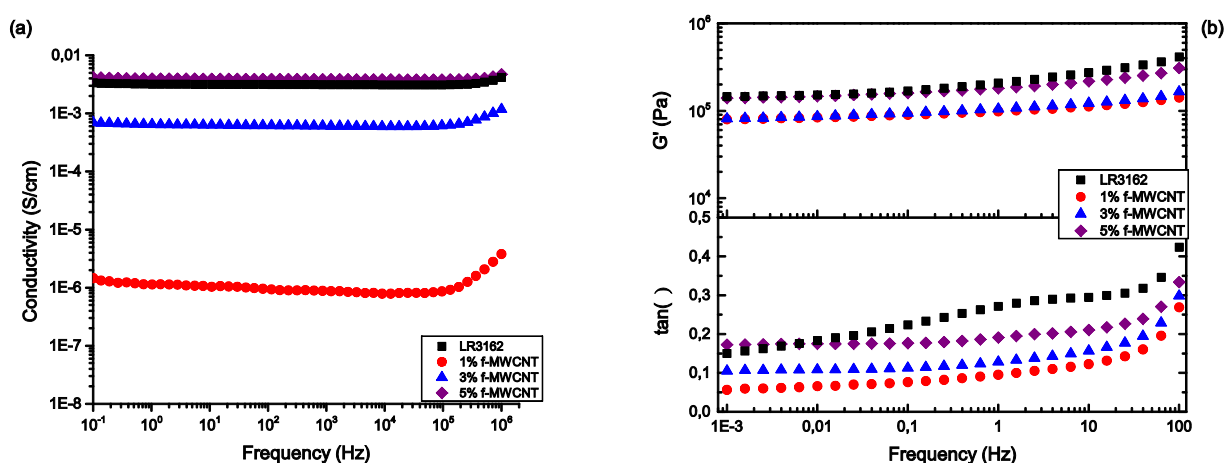
276 A free radical polymerization of MPDMS was conducted in a dispersion of pristine MWCNT in a
 277 mixture of toluene and anisole. The method is very simple to conduct and has the advantage that it
 278 can directly be up-scaled for large amounts of modified MWCNT. The purification process was a
 279 simple precipitation in methanol, which allows both the MWCNT as well as any free polymer
 280 formed to be isolated, while excess monomer will be removed. As shown in Figure 8-left the
 281 amount of grafted polymer on the MWCNT could be determined from thermogravimetric analysis
 282 (TGA).



283 **Figure 8:** TGA of the pristine MWNCT, rinsed f-MWCNT and the unpurified raw product (f-MWCNT) that was used
 284 directly for the composite preparation (a); IR spectrum of the raw product showing the main peaks from PDMS (b).
 285

286 The free radical process results in a combination of free poly(MPDMS) and surface grafted
 287 poly(MPDMS)-MWCNT (f-MWCNT). By rinsing the precipitated raw product with toluene, it was
 288 possible to remove the excess free polymer, as seen from the significantly lower weight loss
 289 observed in TGA. This rinsed product still shows a weight loss compared to the pristine MWNCT,
 290 which proves the polymer is covalently bound to the MWCNT (8 wt% polymer). The rinsed f-
 291 MWCNT is a hard and brittle material of similar appearance to pristine MWCNT, whereas the raw
 292 product is a well dispersed greasy solid. The raw product contained 7 wt% MWCNT, while the
 293 remaining sample mass is either free or covalently bound poly(MPDMS). The polymer structure

294 was confirmed by FT-IR (Figure 8b), where the clear methacrylate stretch at 1728 cm^{-1} in
 295 combination with the PDMS stretches at 1100 , 1015 and 787 cm^{-1} and the absence of the
 296 methacrylic double bond confirmed the structure of the polymer. The prepared raw product is a
 297 homogeneous black, viscous material, which could be used directly after isolation for preparation of
 298 PDMS networks (concentration is given as the amount of MWCNT in the elastomer). The
 299 functionalized materials required much lower intensity dispersion methods and were just mixed
 300 with PDMS prepolymers in a speedmixer and afforded elastomers with conductivities as shown in
 301 Figure 9a.

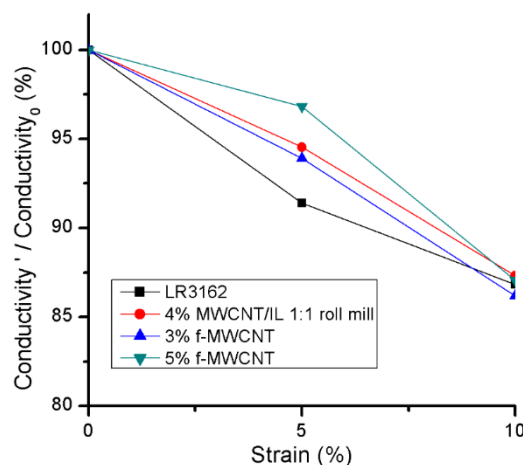


302 **Figure 9:** PDMS composites containing 1-5 wt% f-MWCNT. a) Conductivities at 20°C ; b) Linear viscoelastic response
 303 at 20°C .

304 The functionalized material results in a direct current plateau 2 orders of magnitude above that of
 305 pristine MWCNT and 1 decade above that from the IL-gel procedure for 1wt% MWCNT (Figure
 306 2a). In addition to this, the onset of the plateau is at much higher frequencies for the f-MWCNT
 307 composites, compared to the pristine MWCNT and the MWCNT/IL composites, indicating a very
 308 efficient dispersion. Increasing the content of the f-MWCNT to 3 and 5 wt% resulted in
 309 conductivities on the order of the commercial LR3162. The materials show elastic properties
 310 comparable to that of LR3162 (Figure 9b), with a slightly lower G' for the f-MWCNT at 1 and

311 3 wt%. The losses are significantly lower than for the IL-gel dispersed MWCNT as well as for
 312 LR3162. The combination of both a low G' as well as low losses shows that the network integrity
 313 has been maintained. The functionalization also had a very positive effect on mixing and handling,
 314 where these materials readily disperses in solvent or the PDMS pre-polymer matrix by manual
 315 mixing.

316 Finally, the impact on conductivity by extension of the elastomers for the f-MWCNT and the
 317 MWCNT/IL 1:1 roll mill dispersed samples was compared to the commercial LR3162 at different
 318 strains as shown in Figure 10.



319

320 **Figure 10:** Change in conductivity as a function of strain for the most promising elastomer candidates plotted relative
 321 to the respective original conductivities measured at 20°C.

322 From the plot of relative conductivity of the elastomers it can be seen that the f-MWCNT as well as
 323 the IL dispersed MWCNT retain their initial conductivity to a greater extent than the commercial
 324 reference at 5% strain, whereas all of the samples show an 13 % reduction in conductivity at 10%
 325 strain. The stretchability of particularly the 5 wt% f-MWCNT shows a promising potential for this
 326 type of material at small strains, where there is a significant improvement compared to the reference
 327 material. This is particularly important for applications in joints and connections where a constant
 328 conductivity of such a flexible material is required.

329 **Conclusion**

330 The use of IL/MWCNT-gels as a pre-dispersion method was evaluated for the preparation of highly
331 filled MWCNT/PDMS composites. IL-gels were shown to be effective in dispersing the MWCNT,
332 but the method was not sufficiently robust at higher loading of MWCNT. In order for the method to
333 be applicable it is required that a high reproducibility as well as an easy handling can be obtained.
334 The method allowed the preparation of highly conductive MWCNT/PDMS composites with 4 wt%
335 MWCNT (at a MWCNT/IL 1:1 ratio) but the amount of manual work involved in the preparation
336 was deemed not to give a significant high return in the form of conductivity compared to the other
337 processes. SEM analysis of the elastomers showed that the observed reduction in conductivity at
338 higher amounts of MWCNT could be attributed to a change in morphology occurring between 4
339 and 5wt% MWCNT (or at 4wt% MWCNT with high IL load).

340 When the IL method was combined with a roll mill the dispersion was more effective and more
341 robust. But this also resulted in a significant softening of the network and significant losses in the
342 material. However, under these conditions also pristine MWCNT could be dispersed to a similar
343 degree by combination of a speedmixing step followed by treatment in the roll mill.

344 As an alternative to the roll mill a novel type of PDMS grafted f-MWCNT was prepared and tested
345 using speedmixing as dispersion method. The f-MWCNTs were easier to handle and facilitated a
346 stable dispersion in PDMS pre polymers and composites with conductivities comparable to LR3162
347 without the requirement of the roll mill treatment. The f-MWCNT procedure is an additional step,
348 but the approach is a simple and up-saleable process without the requirement of rigorous and time
349 consuming purification steps.

350 Finally, the conductivity of the elastomers during extension was confirmed up to 10% strain for
351 selected samples, showing a promising improvement in retained conductivity at 5% strain and
352 comparable results at 10% strain compared to LR3162.

353 Overall the approaches illustrate that in order to obtain a good dispersion of MWCNT in PDMS and
354 obtain a high conductivity of the elastomer a sequence of both dispersion aids as well as effective
355 mechanical mixing methods are required. Either speedmixing/US in combination with a roll mill or
356 f-MWCNT in combination with speedmixing was identified as the most effective approaches to
357 obtain highly conductive PDMS composites with maintained mechanical integrity.

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